On the Molecular-Aerosol Scattering Coupling in Remote Sensing of Aerosol From Space

V. V. Rozanov and A. A. Kokhanovsky

Abstract—This work is devoted to studies of the influence of the molecular-aerosol coupling on the scalar approximation-based aerosol satellite remote sensing procedures in the ultraviolet–near-infrared spectral range. It was found that the coupling error must be accounted in the aerosol remote sensing problems based on the analysis of the backscattered ultraviolet light.

Index Terms—Radiative transfer, remote sensing.

I. INTRODUCTION

AEROSOL remote sensing from space over land is often performed assuming that the satellite signal can be decoupled in two components (namely, molecular and aerosol scattering contributions). Then the lookup tables approach is used to retrieve aerosol optical thickness τ_a. Such decoupling would be an exact procedure as the atmospheric optical thickness $\tau_{\text{atm}} \ll 1$. Then scattering by molecules and aerosol particles becomes independent. However, terrestrial atmosphere is a multiply scattering medium as far as ultraviolet (UV)–visible–near-infrared (IR) radiative transport is concerned. It means that the total atmospheric reflection function R can be presented as

$$R = R_m + R_a + \Delta R \tag{1}$$

where R_m is the Rayleigh reflection function defined as the atmospheric reflection function for the hypothetic atmosphere having only gases and no scattering particles, R_a is the aerosol reflection function defined as the atmospheric reflection function for the hypothetic atmosphere having only particles and no gases. For clarity of considerations, we neglect the ground reflection contribution. However, this could be easily taken into account as an additional term in (1).

The value ΔR accounts for a coupling between molecular and aerosol scattering. Equation (1) allows to present the relative error of decoupling process as

$$\varepsilon = \frac{\Delta R}{R}. \tag{1}$$

Calculations show that the value of ε is rather small for typical atmospheric and satellite observation conditions (usually below 3% to 5%). However, for aerosol remote sensing applications, not coupling error ε itself but rather the relative error δ of the

aerosol reflection function determination (in the assumption that coupling can be neglected) is of importance. It follows that

$$\delta = \frac{R_a - R_a^*}{R_a} \tag{3}$$

where $R_a = R - R_m - \Delta R$ and $R_a^* = R - R_m$. We have from (3)

$$\delta = -\frac{\varepsilon}{1 - \varepsilon - \rho} \tag{4}$$

where $\rho = R_m/R$. Clearly, the value of ρ decreases with λ and can be neglected for large wavelengths at most sun/satellite geometries (e.g., in IR). Then it follows approximately: $\delta \approx -\varepsilon$. It means that δ is small if ε is small under assumption $\rho \to 0$. Therefore, δ and ε are of about the same magnitude but of a different sign for small ρ. However, the value of ρ increases toward the UV region and can be in principle considerably larger than the coupling error ε. In particular, it follows from (4) that $\delta \gg -\varepsilon$ as $\rho \to 1$.

Let us imagine the situation, when the atmospheric reflection function is almost entirely determined by molecular scattering processes. Then we have $\rho = 1 - \sigma$, where σ is a small number. Therefore, it follows from (4)

$$\delta = \frac{1}{1 - \nu} \tag{5}$$

where $\nu = \sigma/\varepsilon$. It means that $\delta \to \infty$ as $\nu \to 1$. Therefore, the error of the aerosol reflection function determination neglecting coupling could be quite large if $R \approx R_m$. The coupling errors themselves could be very small in this case. Then we need to detect a small signal on the much higher level background contribution. This explains the increase in the error for this case.

The task of this paper is to study errors ε and δ for realistic atmospheric and illumination/observation conditions. This will allow us to understand if one can neglect the coupling contribution for aerosol remote sensing purposes (e.g., for the determination of the aerosol optical thickness from space). The approximation assuming $\Delta R = 0$ looks quite attractive taking into account that the gaseous composition of atmosphere is very stable. This means that R_m can be calculated quite accurately in advance for a given sun/satellite geometry using available information on the spectral molecular optical thickness [2]. Then the value R_a is approximated as $R - R_m$, where R is the measured reflection function. Lookup tables of precalculated aerosol reflection functions can be searched then to find the correspondent aerosol optical thickness [12].

Similar studies have been performed by Antonie and Morel [1] using Monte Carlo calculations at wavelengths larger than

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TABLE I
AEROSOL VERTICAL MODEL USED IN CALCULATIONS. OPTICAL MODELS
FOR EACH AEROSOL TYPE WERE TAKEN FROM HESS *et al.* [3] AND
SCIATRAN DATABASE [7]. THE SATELLITE SIGNAL IS MOSTLY
INFLUENCED BY THE LOWER AEROSOL LAYER

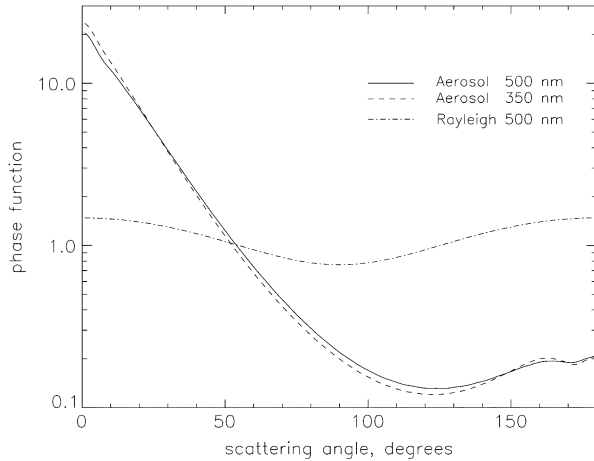| The position of the aerosol layer	Type of aerosol
0-2km	urban
2-10km	average continental
10-30km	sulfate
30-100km	meteoric

Fig. 1. Truncated aerosol phase function (50 Legendre moments are used) at the wavelengths 350 and 500 nm in the lower layer. The dashed–dotted line gives the Rayleigh phase function.

445 nm. They concluded that the coupling between aerosols and molecules contributes significantly to the path reflectance. We extend their study to the case of UV light backscattering.

II. ATMOSPHERIC AND RADIATIVE TRANSFER MODELS

In our radiative transfer simulations of reflection functions R, R_a, R_m (and, therefore, also δ, ε, ν) we use the following atmospheric model. The aerosol optical properties are defined in four layers as shown in Table I. The aerosol singl-scattering albedos, extinction coefficients, and phase functions have been taken from the optical properties of aerosols and clouds (OPAC) atmospheric model [3]. The truncated phase functions for aerosol scattering in the lower atmospheric layer are given in Fig. 1. The truncation is performed according to standard techniques [10]. The use of truncated phase functions speeds up radiative transfer calculations considerably.

Aerosol phase functions differ from the correspondent Rayleigh phase function also given in Fig. 1. In particular, we see that the Rayleigh scattering is much more effective in the backscattering region as compared to the aerosol scattering. This means that purely aerosol and molecular scattering media having the same optical thickness will produce different backscattering signals. Molecular scattering will be by order of magnitude stronger in the single-scattering approximation. The spectral single-scattering albedo for the most contributing to the reflected light layer 1 (see Table I) is shown in Fig. 2. It differs considerably from 1 because of assumed urban
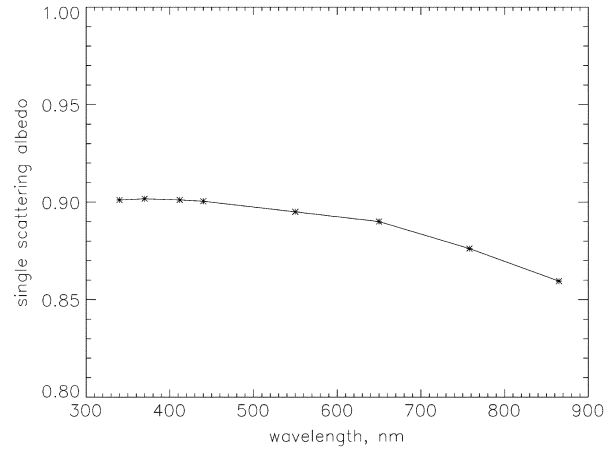

Fig. 2. Spectral single-scattering albedo for the lowest atmospheric layer.

TABLE II
RAYLEIGH OPTICAL THICKNESS AT SEVERAL WAVELENGTHS
USED IN CALCULATIONS [2]

λ, nm	340	370	412	440	550	650	758	865
τ_r	0.714	0.500	0.319	0.243	0.097	0.049	0.026	0.016

soot-polluted aerosol model used in calculations. Because the OPAC model is exclusively based on Mie theory, possible effects of nonspherisity of aerosol particles are neglected. The aerosol optical thickness τ_a (550 nm) was varied in the range 0.05–1.0. The Rayleigh optical thickness have been calculated assuming the 1962 US standard atmosphere model with values of Rayleigh optical thickness τ_r given in Table II. Gaseous absorption was neglected. Calculations were performed at wavelengths 340, 370, 412, 440, 550, 650, 758, and 865 nm using the SCIATRAN 2.0 radiative transfer model developed at Bremen University [7]. SCIATRAN 2.0 is based on the discrete ordinate technique to solve the scalar radiative transfer equation for a plane-parallel turbid layer with account for the vertically varying molecular, aerosol, and cloud light scattering. The program also incorporates databases of atmospheric air local optical characteristics, including gaseous absorption cross sections and concentrations profiles. Further details are given by Rozanov *et al.* [7]. The code and correspondent guide are freely available at http://www.iup.physik.uni-bremen.de/sciatran.

III. RESULTS OF NUMERICAL CALCULATIONS

We show the aerosol reflection function R_a (solid line) calculated with account for coupling as compared to the aerosol reflection function R_a^* (dashed line), calculated in the assumption $\Delta R = 0$ (no coupling) in Fig. 3. Calculations were performed at the spectral points as indicated in Table II at the aerosol optical thickness $\tau_a(550 \text{ nm}) = 0.3$, the satellite zenith angle $\theta = 30°$, and the solar zenith angle $\theta_0 = 60°$ for azimuths ϕ equal to $0°, 90°$, and $180°$. It follows that R_a^* (no coupling) differs from R_a considerably in visible and UV. In particular, the spectral aerosol reflection function derived in the assumption that there is no coupling takes a maximum around 440 nm and then starts to decrease. Such a behavior can be interpreted as an increased
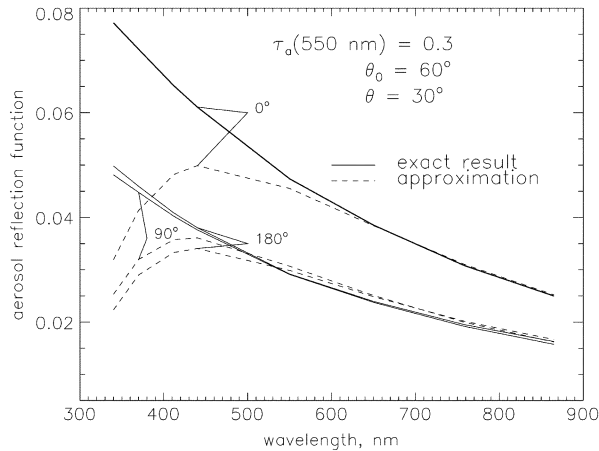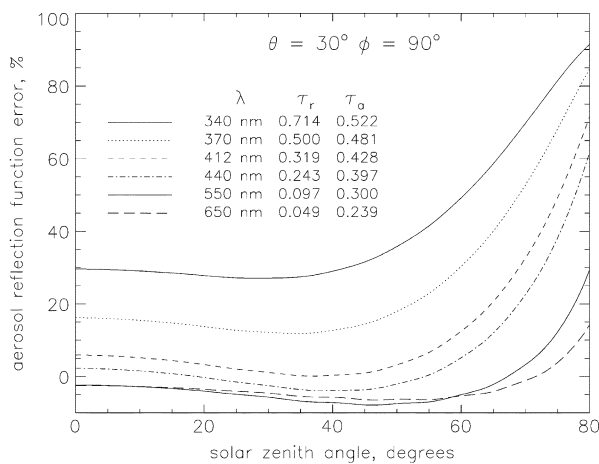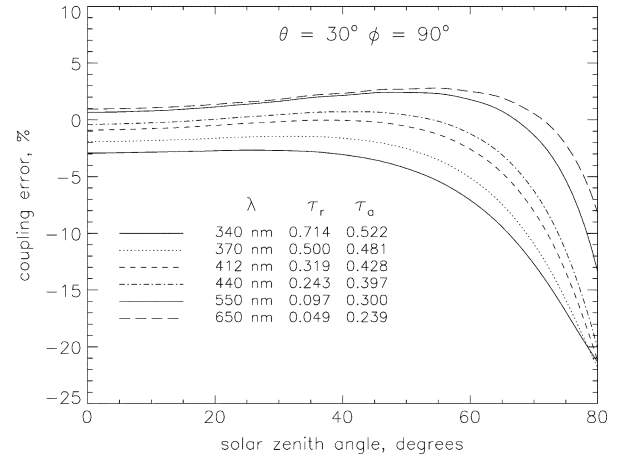
Fig. 3.   Retrieved aerosol reflection function.

Fig. 4.   Dependence of the aerosol reflection function error δ on the solar zenith angle θ_0.

Fig. 5.   Same as in Fig. 4 except for the coupling error ε.

aerosol absorption in UV. This is in contradiction with the model used (see Fig. 2) and also with experimental results (e.g., see [9] and [12]). So clearly the coupling between molecular and aerosol scattering cannot be neglected in the satellite retrievals algorithms using spectral range below 440 nm (and even below 500 nm depending on the geometry; see Fig. 3). This is an unfortunate situation taking into account that the reflective power of most natural surfaces is much smaller in UV as compared to visible. So the UV spectral range is very attractive for aerosol remote sensing as compared to retrievals in visible due to the minimization of the generally unknown surface influence error [5], [11], [13].

The angular dependence of the aerosol reflection function error δ is shown in Fig. 4 at $\theta = 30°, \phi = 90°, \tau_a(550\,\mathrm{nm}) = 0.3$ and wavelengths 340, 370, 412, 440, 550, and 650 nm. It follows that the error increases considerably with the solar angle and the inverse wavelength. It is around 30% at the wavelength 340 nm for the solar zenith angles below 40°. However, it sharply increases for low sun producing the positive error of about 90%. It means that the retrieved value of the aerosol reflectance R_a^* under the assumption of no coupling will be too low. This will lead to a considerable underestimation of the aerosol optical thickness. The error is smaller at the wavelength 412 nm and solar angles smaller than 60°. It is below

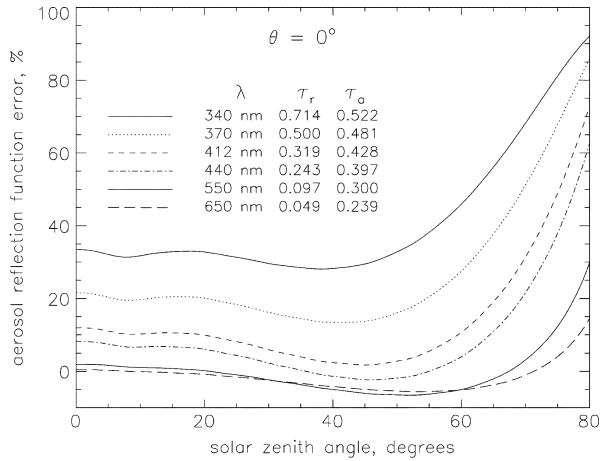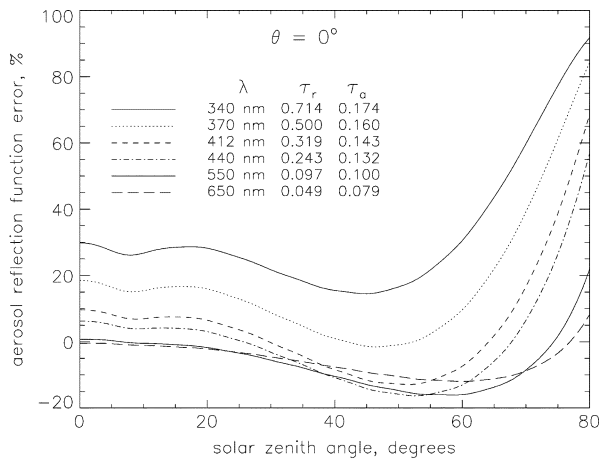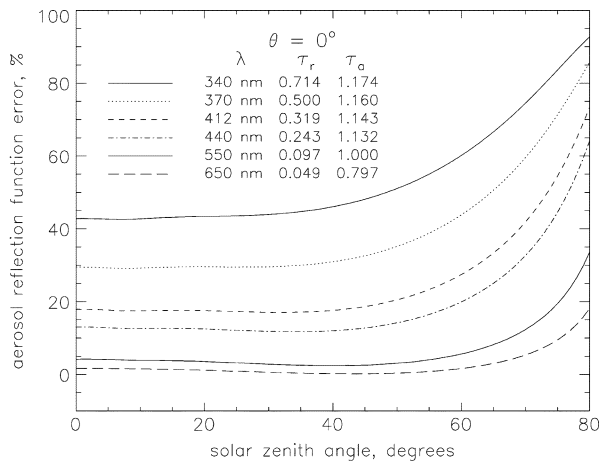10% and sun-position dependent then. The error is negative and small (typically, below 5%) at wavelengths larger 550 nm and solar zenith angles usually used in the aerosol remote sensing ($\theta_0 < 70°$). Note that aerosol retrievals over oceans are often performed in IR channels (e.g., 865 nm) due to the small contribution of light reflected from ocean in this case (outside of the glint region). It is fortunate that the coupling error may be neglected for maritime aerosol remote sensing performed at $\lambda > 700$ nm. However, even in this most favorable case, the error can reach 15% at the solar angle 80° and the wavelength 650 nm. This could affect the quality of retrievals in polar regions, where sun is low. The errors are negligible for wavelengths 758 nm, 865 nm due to low molecular scattering contribution in this case (see Table II).
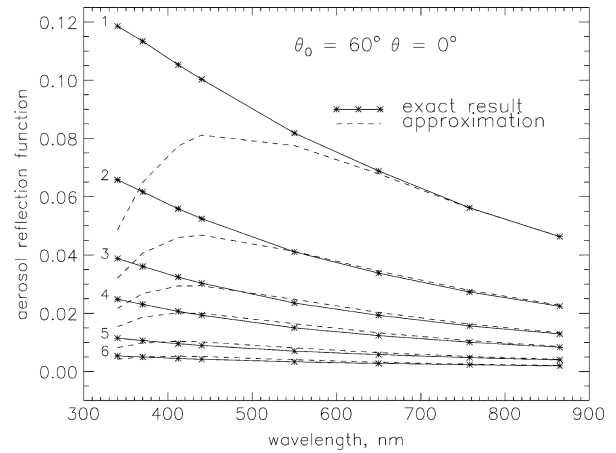
Note that the coupling error ε itself is not so large in the cases considered (see Fig. 5). The increase of errors toward lower sun positions is best explained by the fact that light needs to travel larger distances in the atmosphere in this case. This rises up the molecular scattering contribution. Indeed, the molecular optical depth differs considerably along vertical and along slant paths in the atmosphere. This is the reason why ε is below 1% at the solar zenith angle 0° but around -8% at $\theta_0 = 80°$ (see Fig. 5). The coupling error is mostly negative suggesting that $\Delta R < 0$ in most of cases and, therefore, if one neglects coupling errors, too small aerosol reflectances are obtained and, therefore, the retrieved aerosol optical thickness is underestimated. This is consistent with Fig. 4, where $\delta > 0$ for most of cases. Also it follows from Figs. 4 and 5: $\delta \approx -\varepsilon$ at small ε as it was suggested above.

Results considered above were calculated for the observation angle 30° and the aerosol optical thickness $\tau_a(550\,\mathrm{nm}) = 0.3$. We have decided to check how errors depend on the observation geometry and aerosol optical thickness. Therefore, we have repeated our calculations for other observation geometries, and values of τ_a. Selected results of this study are shown in Figs. 6–8. Results given in Fig. 6 have been calculated at the same conditions as in Fig. 4, but the observation angle was changed. We see that our conclusion in respect to importance of coupling term ΔR is not influenced very much by the observation geometry change. So they are of general nature.

The influence of the aerosol load is illustrated in Figs. 7 and 8, where results for $\tau_a(550\,\mathrm{nm}) = 0.1$ and $\tau_a(550\,\mathrm{nm}) = 1.0$

Fig. 6.   Same as in Fig. 4 except at $\theta = 0°$.

Fig. 7.   Same as in Fig. 6 except at $\tau_a(550\,\text{nm}) = 0.1$.

Fig. 8.   Same as in Fig. 6 except at $\tau_a(550\,\text{nm}) = 1.0$.

Fig. 9.   Retrieved aerosol reflection function at $\tau_a(550\,\text{nm}) = 0.05(6)$, $0.1(5), 0.2(4), 0.3(3), 0.5(2), 1.0(1)$.

properly accounted for. The underestimation is even larger at $\theta_0 > 60°$, which makes coupling terms even more important in this case. This conclusion is of special importance for arctic aerosol studies although situations with $\tau_a(550\,\text{nm}) = 1.0$ are not frequent there. Fig. 7 shows that the coupling term is still of importance even at $\tau_a(550\,\text{nm}) = 0.1$ for the solar angles larger than $70°$ and $\lambda \leq 440$ nm. Oscillations on Fig. 7 are due to the influence of correspondent oscillations in the phase function (see Fig. 1). Note that the scattering angle at the nadir observation is equal to $\pi - \theta_0$.

Fig. 9 is similar to Fig. 2 but for another observation conditions and several aerosol optical thicknesses. It confirms that the assumption $\Delta R = 0$ leads to unphysical maximum in the retrieved aerosol reflectance positioned at $\lambda \approx 440$ nm and generally smaller values of the aerosol reflection function as compared to true (solid lines in Fig. 9) ones.

Our results are obtained using the scalar approximation of the radiative transfer theory. We also estimated the errors related to the vector character of light not accounted for in our model. For this we have used the vector code SCIAPOL developed by us [8]. Polarization effects can be neglected at $\lambda \geq 650$ nm for Rayleigh scattering and for all wavelengths considered for aerosol scattering. However, they are of importance for smaller wavelengths for Rayleigh scattering and highly solar angle-dependent [6]. In particular, we found (not shown here) that errors in the Rayleigh scattering component due the scalar approximation are in the range 0% to 10% depending on the solar angle in the interval of solar angles $\Delta\theta_0 \in [0°, 50°]$, $\lambda = 340$ nm and the nadir observation [8]. The error is smaller for larger solar angles in the interval $\Delta\theta_0$.

It means that instead of (1) we actually have

$$R = R_m + R_a + \Delta R + \Delta R_v \qquad (6)$$

where ΔR_v is the contribution to the total reflectance due to the vector properties of light. It also means that ε, δ should be substituted by

$$\varepsilon^* = \frac{\Delta R + \Delta R_v}{R}, \quad \delta^* = -\frac{\varepsilon^*}{1 - \varepsilon^* - \nu} \qquad (7)$$

where the star means that the correspondent error is due to fact that both coupling and vector nature of light are not accounted

are given. Calculations were done at the same conditions as in Fig. 6. Again we see that the coupling term cannot be neglected in UV. The value of δ varies in the range 20% to 90% at $\lambda = 340$ nm depending on τ_a and θ_0. Dependencies on θ_0 are monotonous at $\tau_a(550\text{nm}) = 1.0$ because of greater influence of multiple scattering in this case. Errors are generally larger then. They are around 20% at the wavelength 412 nm and $\theta_0 \leq 60°$. This will produce the underestimation of the retrieved aerosol optical thickness $\tau_a(412\text{ nm})$ if the coupling is not
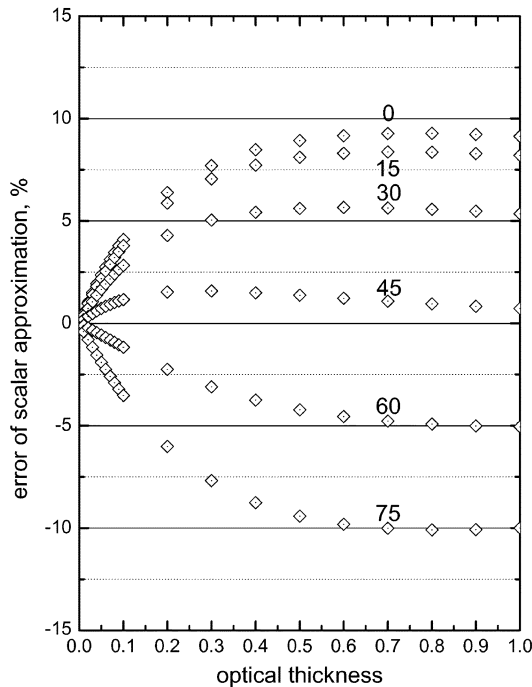
Fig. 10.   Dependence of the scalar approximation error on the optical thickness for Rayleigh scattering at no absorption and various solar illumination angles and the nadir observation.

for. The value of δ is positive (see Fig. 8) at $\tau_a = 1.0$. This means that both $\varepsilon = -(1-\rho)\delta/(1-\delta)$ and ΔR are negative. We found that the same is true for ΔR_v at $\theta_0 \geq 50°$ (see Fig. 10). This will lead to larger absolute values of the pair $(\delta^*, \varepsilon^*)$ as compared to the absolute values of ε and δ and at large solar angles.

Note that it follows: $\Delta R_v > 0$ at $\theta_0 \leq 45°$ (see Fig. 10). In this case the absolute values of the pair $(\varepsilon^*, \delta^*)$ are smaller as compared to the absolute values of ε and δ due to the partial cancellation of errors. This will induce important differences if different algorithms are used for the same radiance field interpretation (e.g., between results obtained using aerosol retrieval algorithms in scalar and vector modes at $\Delta R = 0$). The main conclusion is that not only the coupling error but also the polarization effects should be considered in aerosol remote sensing applications in UV (e.g., see [5]).

UV remote sensing of atmosphere over land is favorable as compared to visible-NIR techniques. Then generally unknown contribution from the surface is small. However, UV aerosol remote sensing algorithms suffice from the necessity to account for the vector nature of light and increased Rayleigh contribution. The latter depends on the ground elevation. Therefore, correspondent UV retrieval techniques become more complex. This is the balanced, however, by much higher accuracy of the UV aerosol optical thickness product as compared to τ_a obtained using visible/NIR retrievals techniques, where light reflected from ground makes large and generally unknown contribution to the satellite signal.

IV. CONCLUSION

We have shown that the coupling of molecular-aerosol scattering is an important issue of the aerosol remote sensing over

land, where wavelengths smaller than 440 nm are used to reduce the uncertainty related to the ground reflectance characteristics. In particular, the retrieved aerosol optical thickness is underestimated in this case. Also a wrong absorption-like feature is introduced in the retrieved aerosol reflectance and the aerosol optical thickness. This may lead to errors in the retrieved aerosol single-scattering albedo, which is of importance for climate problem. Results were obtained in a number of simplified assumptions like the spherical shape of particles, no ground reflection, and the scalar approximation of the radiative transfer. However, main results do not change very much even we account for these factors. In particular, nonspherical particles increase lateral scattering. This will lead to the increase of the aerosol phase function. This will make this function closer to that of Rayleigh scattering thereby strengthening coupling effects. We also estimated the errors related to the vector character of light not accounted for in our model. These effects can be neglected at $\lambda \geq 650$ nm. However, they are of importance for smaller wavelengths as discussed in the text.

One of the important conclusions of this work is that for retrieval of aerosol properties from space for wavelengths below 440 nm, the iterative vector algorithms as those discussed by Hosekamp and Landgraf [4] should be preferably used. The use of *a priori* assumptions on the scalar approximation/strength of coupling are not needed then.

We also show the capabilities of SCIATRAN and SCIAPOL radiative transfer codes in the solution of this important atmospheric optics problem. The SCIAPOL_1.0 radiative transfer code can be downloaded for noncommercial use at www.iup.physik.uni-bremen.de/~alexk. The SCIATRAN_2.0 code is located at www.iup.physik.uni-bremen.de/sciatran.

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